Negative refractive index response of weakly and strongly coupled optical metamaterials.

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Metamaterials are artificially engineered structures that have properties, such as negative refractive index, n, nonexistent in natural materials. The recent development of metamaterials [1–3] with negative n confirms that structures can be fabricated and interpreted as having both a negative permittivity, ϵ, and a negative permeability, μ, simultaneously. Since the original microwave experiments for the demonstration of negative index behavior in split ring resonators (SRRs) and wire structures, new designs have been introduced, such as fishnet, that have pushed the existence of the negative refraction at optical wavelengths [4–9]. Most of the experiments with the fishnet structure measure transmission, T, and reflection, R, and use the retrieval procedure [10–13] to obtain the effective parameters, ϵ, μ, and n. Although, stacking of three [14], four [15], 10 [16] functional layers and recently fabricated [9] 10-functional layer fishnets (21 layers of silver and MgF_2) have been realized, they do not constitute a bulk metamaterial. Even the thickest fabricated [9] fishnet structure only has a total thickness, 830 nm, half of the wavelength (λ=1700 nm). Here, we report a detailed study of the weakly and strongly coupled fishnets to understand the origin of negative n, as well the mechanism of low losses (that is, high figure of merit (FOM)) for the strongly coupled fishnets. We also study the convergence of the retrieval parameter (ϵ, μ and n) as the number of unit cells (layers) increases. For the weakly coupled structures, the convergence results for n and FOM are close to the single unit cell. As expected, for the strongly coupled structures, hybridization is observed and the retrieval results for n and FOM are completely different from the single unit cell. We demonstrate that the high value of FOM for the strongly coupled structure is due to periodicity effects.

The idea of left-handed materials, i.e., materials with both negative ϵ and negative μ, where the electric field (**E**), magnetic field (**H**), and wave vector (**k**) form a left-handed coordinate system was developed by Veselago [17] decades ago. However, it was only recently that such materials were investigated experimentally [4–9], and the field is driven by a wide range of new applications, such as ultrahigh-resolution imaging system [18], cloaking devices [19, 20], and quantum levitation [21]. Realizing these applications, several goals must be achieved: three-dimensional rather than planar structure, isotropic design, and reduction of loss.

Most of the metamaterials exhibiting artificial magnetism [15, 16, 22–24] and a negative refractive index, n, at THz and optical frequencies [4–7, 24], consist of only a functional layer. The number of actual layers $M = 2 \times N + 1$, where N is the number of functional layers. The first five-functional-layer of SRRs operating at 6 THz was published [16] in 2005, and four layers of SRRs operating at 70 THz [15] was published in 2008. The first three-functional-layer of fishnets (7 layers of silver and MgF_2) operating at 200 THz was published [14] in 2007, and recently a 10-functional-layer of fishnets (21 layers of silver and MgF_2) operating at 200 THz was fabricated [9]. Even the thickest fabricated [9] fishnet structure has a thickness (830 nm) half of the wavelength (λ =1500 nm). However, it is very important to study how the optical properties (ϵ, μ and n) change as one increases the number of layers. How many layers are needed to achieve convergence of the optical properties and one can call this metamaterial bulk? How do optical properties behave as one changes the distance between two neighboring fishnets? If the distance is small, we have a strong coupling case. The convergence of optical properties is slow, and more importantly, it does not convergence to the isolated fishnet case. What is the mechanism for negative n in the strong coupling limit?

In this letter, we present a detailed study of the retrieved optical parameters, ϵ, μ, and n of the single fishnet metamaterial structures as a function of the size of the unit cell. We find that as the size of the unit cell decreases, the magnitude of the retrieved effective parameters increases. In order to understand the underlying physics of the coupled structures, we study the retrieved parameters of the coupled fishnets as a function of the distance between them. Finally, we study the convergence of the retrieved parameters as the number of the unit cell increases for the weakly and strongly coupled structures. For the weakly coupling case, the retrieved parameters are very close to the one-functional layer results and converge relatively fast. For the strong coupling case, the retrieved parameters are completely different than the one unit fishnet results. The strong coupling case explains the recently observed negative refractive index in the 21-layer fishnet structure [9], especially the high FOM, due to the periodicity effects, as will be shown below.

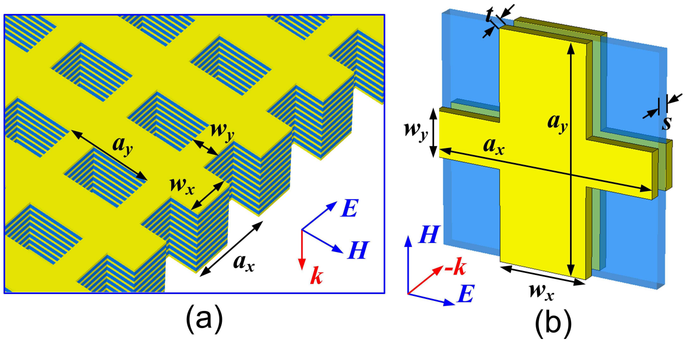

FIG. 1: (a) Schematic of a fishnet structure with 11 metallic layers, (b) a single unit cell with geometric parameters marked on it.

In Fig. 1 we present a schematic graph of the unit-cell of the fishnet structure. The size of the unit cell along the propagation direction is a_z. a_z is larger than the sum of the thickness of the metallic and the dielectric layers $2t + s$, where t and s are the thicknesses of the metal and dielectric layers, respectively. Notice the propagation direction is perpendicular to the plane of the fishnet.
In most of the experiments measuring the T and R of the fishnet structure [4–8, 24], there is only one layer of the sample measured. In this case, the unit cell size along the propagation direction, a_z, is undefined. We have shown [25] that, as a_z decreases, the magnitude of the retrieved parameters increases. It is well known from electronic systems that a monolayer of a surface can exhibit different properties from the bulk (many layers). So it is very important to systematically study whether the optical parameters of a single layer really correspond to the many layers system. We will study the weak and strong coupling limit of the two-layer fishnet structure.

Figure 2 shows the real part of the effective refractive index, Re(n), as a function of λ/a, for one layer and two layers of the fishnet structure described in Fig. 1, with different distances between the unit cells. Notice the normalized resonance wavelength $\lambda_m/a \approx 2.02$, i.e. wavelength with maximum $|\mathrm{Re}(n)|$, for one layer shifts only slightly when the size of the unit cell increases, but the magnitude of $|\mathrm{Re}(n)|$ decreases dramatically. For the two layers, when the distance, d, between them is large (d/a=0.24, blue solid curve), the coupling between the two layers is weak and, therefore, the refractive index, Re(n), approaches the one layer simulation results. When the distance between the two layers becomes smaller (d/a=0.04, red solid curve) and the coupling becomes stronger, hybridization takes place and two resonance modes exist, one at $\lambda/2 = 2.005$, which gives

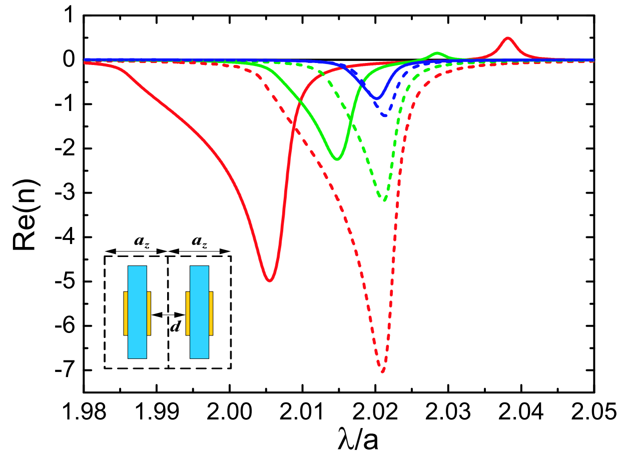

FIG. 2: Retrieved real part of refractive index, n, from simulated data using unit cell size in the propagation direction $a_z = a/15$ (red), $a_z = 2a/15$ (green) and $a_z = 4a/15$ (blue). Both one layer (dashed) and two layers (solid) results are shown. The distances between two unit cells are $d = a_z - (2t + s)$ =0.04a, 0.11a, and 0.24a, respectively. The other geometric parameters are given by $a_x = a_y = a$, $w_x = 4a/15$, $w_y = 3a/5$, $s = a/60$, $t = a/300$, and the dielectric constant of the spacer is $\epsilon_r = 5$.

Re(n) < 0; and one at 2.040, which has Re(n) > 0. The difference in value of the two resonance frequencies becomes larger as the distance between them decreases. Another very important issue is how fast the optical retrieval properties (ϵ, μ and n) converge as the number of unit cells increases. We will present results for two cases, one for the weakly coupled fishnets and one with the strongly coupled fishnets, which represent the recently published results [9] for 21 layers.

The best design that gives negative n at THz and optical frequencies is the so-called "double-fishnet" structure, which consists of a pair of metal fishnets separated by a dielectric spacer [4–9]. For the incident polarization shown in Fig. 1, the thin metallic wires along the x-axis, parallel to the incident electric field, $\mathbf{E}$, excite the plasmonic response and produce negative permittivity ϵ up to the plasma frequency. Negative μ is obtained from the wires along the y-axis, parallel to the incident magnetic field $\mathbf{H}$. At the magnetic resonance frequency, the two parallel bars sustain anti-parallel currents (along x-axis), providing a magnetic field $\mathbf{B'}$, mainly between the plates and directly opposite to the external magnetic field, $\mathbf{H}$. The electric field, because of the opposite charges accumulate at the ends of the two metallic bars, is expected to be confined within the space between the plates and near the end points. Indeed, obtained simulations confirm this picture.

In Fig. 3 we present the retrieved results for the effective refractive index, Re(n), as a function of λ for

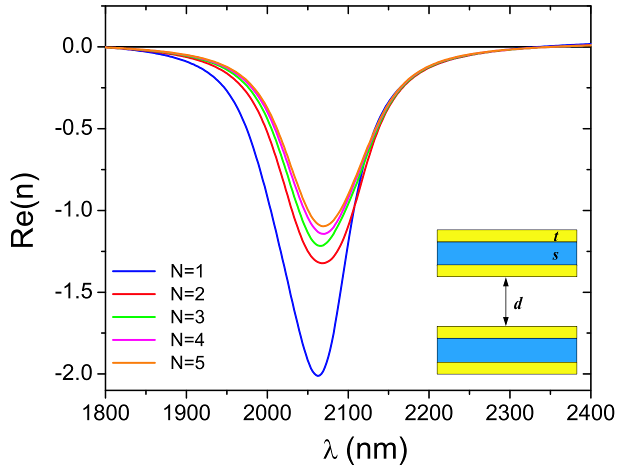

FIG. 3: Retrieved real part of effective refractive index, Re(n) for one layer (red solid), four layers (blue dashed), eight layers (green dotted) and ten layers (black dash-dotted) of the fishnet structure. The geometric parameters are $a_x = a_y = 860$ nm, $w_x = 565$ nm, $w_y = 265$nm, $s = 50$ nm, $t = 30$ nm, $d = 90$ nm, and the spacer is made from MgF_2 with the dielectric constant $\epsilon_r = 1.9$. The functional layers is spaced by vacuum.

different numbers of functional layers (N=1, 2, 3, 4 and 5) for weakly coupled fishnets system. The parameters are exactly the same as the strongly coupled case, that will be discussed below, but the spacing between the functional layers is $d = 90$ nm. As can be seen in Fig. 3, the retrieved results for Re(n) converge very fast (N=2) and the convergence results agree with the results of the one functional layer of the fishnet. When the fishnets

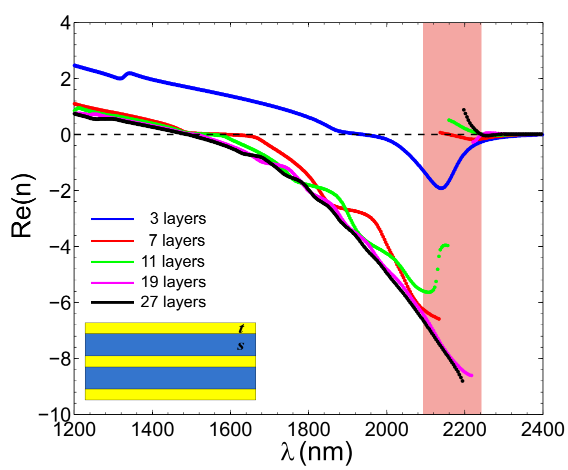

FIG. 4: The retrieved real part of n for 3, 7, 11, 19 and 27 layers strongly coupled fishnet structure. The geometric parameters are $a_x = a_y = 860$ nm, $w_x = 565$ nm, $w_y = 265$nm, $s = 50$ nm and $t = 30$ nm, and the spacer is made from MgF_2 with the dielectric constant $\epsilon_r = 1.9$. The shadow region shows where the discontinuity happens.

strongly interact, it's not clear what the mechanism is for giving negative n. As discussed in Fig. 2, the isolated fishnet resonance frequency hybridizes into two different modes. The antisymmetric mode gives weak resonance with $n \approx 0$, while the symmetric mode gives a strong resonance with a strong negative n. In Fig. 4 we present results for the retrieved, Re(n), for different number of layers (3 to 27) for the recently fabricated [9] negative index structure. Notice in the low wavelength limit (between 1200 to 2100 nm), convergence of n is obtained and agrees with experimental results of Ref. [9]. In the high wavelength limit ($\lambda > 2200$ nm), the Re(n) is zero and the Im(n) is much larger than the Re(n), exhibiting metallic behavior. This metallic behavior can be also seen in the transmission, T, (not shown) for the many layer structure. Above 2200nm, T is low, and behaves as a metal, while for $\lambda < 2100$ nm, T is relatively large (~ 0.8) and has Fabry-Perot resonances structure. In addition, in Fig. 4 the 3-layer structure (the single fishnet structure) gives results completely different than those for the strongly coupled fishnets. These single fishnet results agree with those presented in Fig. 3. Another important quantity is the figure of merit (FOM) which can be defined two different ways. The usual definition is FOM=$|\mathrm{Re}(n)/\mathrm{Im}(n)|$ and the experimental definition of Im(n) is given by $\mathrm{Im}(n) = (\lambda/4\pi d)\ln[(1-|R|)/|T|]$, where λ, d, R, and T are the wavelength, sample thickness, reflectance, and transmittance, respectively.

In Fig. 5 we present the results of the FOM as a

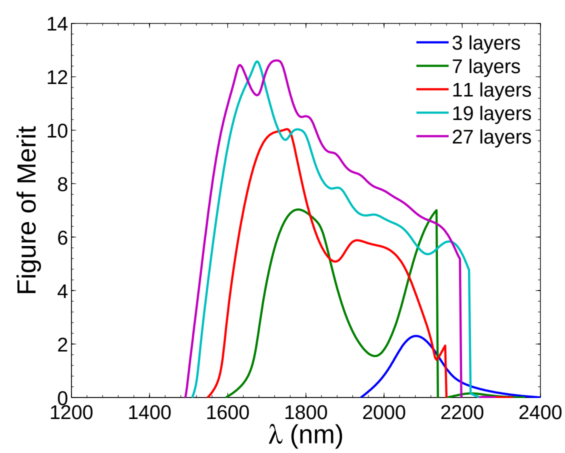

FIG. 5: The figure of merit (FOM) of Re(n) < 0 region for 3, 7, 11, 19, and 27 layers strongly coupled fishnet structure. The FOM is calculated by FOM=$|\mathrm{Re}(n)/\mathrm{Im}(n)|$, where Re($n$) is obtained by a retrieval procedure and Im(n) is calculated by $\mathrm{Im}(n) = (\lambda/4\pi d)\ln[(1-|R|)/|T|]$.

function of wavelength for different number of layers. For the one unit cell fishnet (3 layers), the FOM is really small (of the order of 2) and is located at $\lambda =$ 2100nm, the resonance frequency of the single fishnet structure. As the number of layers increases, the FOM increases and finally saturates to a constant value of the order of 10. This behavior of the FOM for the strongly coupled fishnets is completely different for the weakly coupled fishnets, where the FOM does not change dramatically [25] as one uses more unit cells. Why is the FOM in the strongly coupled fishnets so much different than the single fishnet? It has been argued [9, 26] that the FOM is larger because of the strong coupling between the neighboring layers, which provides destructive interference of the antisymmetric currents across the metal film and effectively cancels the current in the center of the film, and, therefore, reduces the losses. We have systematically studied the current density for the different number of strongly coupled fishnet structures. For the single fishnet structure, the current density is along opposite directions in the two metallic bars. This is the typical behavior of negative index materials. When the number of layers increases, the current density is more complicated and there is no clear physical explanation why one obtains negative n and why the FOM is so large.

In Fig. 6, we present the current density along the

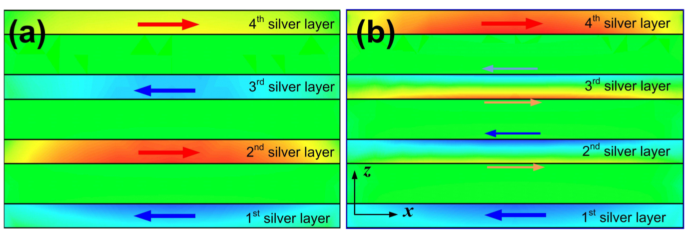

FIG. 6: (a) The current density distribution for a 7 layers strongly coupled fishnet at wavelength, $\lambda = 2230$ nm (antisymmetric mode), with $\mathrm{Re}(n) = -0.17$. (b) The current density distribution for a 7 layers strongly coupled fishnet at wavelength, $\lambda = 1859$ nm (symmetric mode), with $\mathrm{Re}(n) = -2.5$. The cross-section is perpendicular to the y-axis (i.e., incident magnetic filed, $\mathbf{H}$, direction). The color shows the current density in x-direction, J_x, with the red and blue being the positive maximum and negative maximum of J_x, respectively. The arrows show the direction of current density inside the silver layers schematically.

x-axis (or $\mathbf{E}$-direction as shown in Fig. 1), J_x, of the antisymmetric and symmetric modes for the seven layers (4 metallic layers and 3 dielectric layers) strongly coupled structure. For the antisymmetric mode (as shown in Fig. 6(a)), two double-fishnets are formed by the first and second silver layers, and by the third and fourth silver layers. The induced current inside two double-fishnets excite the magnetic fields, $\mathbf{B}'$, along the same direction. However, the second and the third silver layers also form a double-fishnet, which excits the magnetic fields in the opposite direction. Therefore, the excited magnetic fields, $\mathbf{B}'$, are always anti-parallel in the space between neighboring silver layers and cancel each other. This explains the observation of a weak resonance with nearly zero n. For the symmetric mode shown in Fig. 6(b), the first and the fourth silver layers have current density along opposite directions and are almost uniform for all the metallic thickness of 30 nm silver layers. In the second and third silver layers the current density is no longer uniform in all thicknesses of the silver layers. Instead, the current flows along opposite directions on the two surfaces of each layer. Due to the anti-parallel current on the surfaces of the second and third silver layers, the induced magnetic field, $\mathbf{B}'$, in the space between neighboring silver layers, is always parallel to each other. As a consequence, the 7 layers structure can be viewed as three cascade double-fishnet structures with the induced magnetic fields, $\mathbf{B}'$, along the same direction. Therefore, the symmetric mode results in a strong resonance with large negative n. It is not clear, this current density distribution is responsible for the low losses and high FOM [9, 26]. We believe the low losses and high FOM are due to the periodicity effects [11].

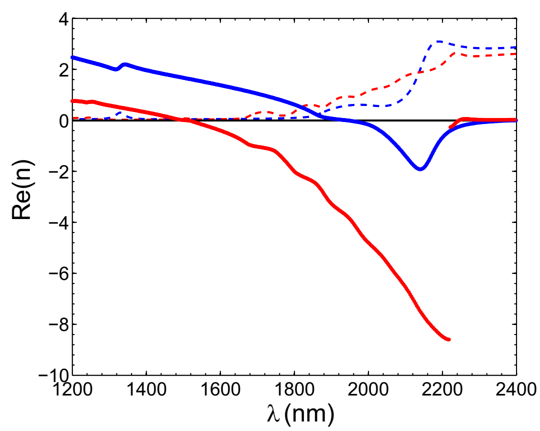

FIG. 7: The real (solid curves) and imaginary (dashed curves) parts of refractive index, $\mathrm{Re}(n)$ and $\mathrm{Im}(n)$, for the 3 and 19 layers fishnet structures. The black dash line shows the position where $\mathrm{Re}(n) = -1$.

In Fig. 7, we present both the real and the imaginary parts of the refractive index, $\mathrm{Re}(n)$ and $\mathrm{Im}(n)$, for the 3 and 15 layers fishnet structures. For the 3 layers (the single layer of double-fishnet), the $\mathrm{Re}(n)$ has a smooth resonance curve (blue solid). The bandwidth of the $\mathrm{Re}(n) < 0$ region is relative narrow and close to the peak of $\mathrm{Im}(n)$ (blue dashed), so the figure of merit is very small (as shown in Fig. 5). For the 15-layer fishnets, the $\mathrm{Re}(n)$ curve (red solid) does not have the resonance behavior expected for a single functional layer, but it's very broad and has structure which is due to periodicity effects [11]. Notice that for the 15 layer structure $\mathrm{Re}(n)$=-1 at $\lambda = 1750$ nm and the $\mathrm{Im}(n)$ is 0.14, so the FOM is of the order of 10. However, for the 3 layer structure, Re(n)=-1 at $\lambda = 2075$ nm and 2185 nm, and the Im(n) is 0.44 and 1.43 respectively, so the FOM is of the order of 1. Therefore, due to the distortion of Re(n) caused by the periodicity effects, the FOM of the fishnet structure increase dramatically as the number of layers increases.

We have made a systematic study of the weakly and strongly coupled fishnets to understand the origin of negative n, as well as the origin of losses and the large value of the FOM for the strongly coupled fishnets. We studied the size dependence of the retrieved parameters (ϵ, μ, and n) of the weakly and strongly coupled fishnet structures. For both cases we found the retrieved parameters have a strong resonance behavior as the size of the unit cell decreases. We have also studied the convergence of the retrieved parameters, as the number of unit cells (layers) increase. For the weakly coupled fishnet structures, we found the convergence results are relatively close to the single unit cell. Also, the converged FOM for the weakly coupled fishnet is the same order of magnitude as the single fishnet. For the strongly coupled fishnet structures, we demonstrated that hybridization happens and we have two resonance modes. The antisymmetric resonance mode gives negative n. As more unit cells or layers are added, the convergence of the retrieval parameters are completely different than the single fishnet results and the FOM is much larger than the single fishnet. We have demonstrated that the large FOM for the strongly coupled fishnet is due to the periodicity effects. Our converged results for the negative index of refraction, n, agree very well with the recent experiments of the strongly coupled fishnet.

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